

Reconciling Semiclassical and Bohmian Mechanics:

I. Stationary states

Bill Poirier

*Department of Chemistry and Biochemistry, and Department of Physics,
Texas Tech University, Box 41061, Lubbock, Texas 79409-1061**

The semiclassical method is characterized by finite forces and smooth, well-behaved trajectories, but also by multivalued representational functions that are ill-behaved at caustics. In contrast, quantum trajectory methods—based on Bohmian mechanics (quantum hydrodynamics)—are characterized by divergent forces and erratic trajectories near nodes, but also well-behaved, single-valued representational functions. In this paper, we unify these two approaches into a single method that captures the best features of both, and in addition, satisfies the correspondence principle. Stationary eigenstates in one degree of freedom are the primary focus, but more general applications are also anticipated.

I. INTRODUCTION

Theoretical and computational physical chemists have long sought reliable and accurate means of performing quantum dynamics calculations for molecular systems, as quantum effects such as tunneling and interference often play an important role in such systems. If “exact” methods are required—i.e., numerical techniques for which the error bars may (in principle) be reduced arbitrarily—the traditional approach has been to represent the system Hamiltonian using a finite, direct-product basis set. However, this approach suffers from the drawback that the scaling of computational effort is necessarily exponential with system dimensionality.^{1,2,3}

Recently, a number of promising new methods have emerged that may spell an end to exponential scaling—or at the very least, drastically reduce the exponent. The latter category includes various basis set optimization methods,^{4,5,6,7,8} which have nearly doubled the number of degrees of freedom (DOFs) that may be tackled on present-day computers, from about 6 to 10 DOFs. Very recently, the first basis-set method to defeat exponential scaling (at least in principle) was introduced.^{9,10,11} This method, which uses wavelets in conjunction with a phase space truncation scheme, has been applied to model problems up to 15 DOFs, and is easily extendible to higher dimensionalities—although some technical issues vis-a-vis applicability to real molecular systems must still be resolved.

A completely different approach to the exact quantum dynamics problem may be found in time-dependent trajectory methods. Although trajectory methods are extremely common in molecular dynamics applications, they are almost always classical, quasiclassical, or semiclassical^{12,13,14,15,16,17,18,19}—i.e., not exact, in the sense described above. However, it is possible to perform exact quantum dynamical propagation using trajectory-based methods. These so-called “quantum trajectory methods”^{20,21,22,23,24} (QTM) are based on the hydrodynamical picture of quantum mechanics, developed over half a century ago by Bohm^{25,26} and Takabayasi,²⁷ using even earlier ideas of Madelung¹² and van Vleck.¹³

Trajectory methods of all kinds are appealing, because they offer an intuitive, classical-like understanding of the underlying dynamics. QTMs are especially appealing, however—not only because they ultimately yield exact results, but also because they offer a pedagogical understanding of quantum effects such as tunneling.^{20,24} Curiously, QTMs thus far have not fared so well as semiclassical methods, vis-a-vis their treatment of another fundamental quantum effect—interference. This issue is discussed in more detail below, as it is of central concern for the present paper. An in-depth comparison of interference phenomena is provided in an intriguing article by Zhao and Makri.²⁸ Garashchuk and Rassolov^{29,30} discuss the interesting connection between QTM and semiclassical propagators,^{15,17,18} in the context of Herman-Kluk initial value representations.^{31,32,33}

Perhaps the greatest attraction of QTMs, however, has been the promise that they may be able to defeat exponential scaling. In any event, QTMs have undergone tremendous development since their introduction in 1999—most notably within the last year or two. Much of the early development centered around accurate evaluation of spatial derivatives of the wavefunction,^{20,21,34} but with the introduction of local least-squares fit adaptive and unstructured grid techniques,^{22,23,24} this difficulty is now essentially resolved. This has paved the way for a number of interesting applications of QTMs, including barrier transmission,²⁰ non-adiabatic dynamics,³⁵ and mode relaxation.³⁶ Several intriguing phase space generalizations have also emerged,^{27,37,38,39,40} of particular relevance for dissipative systems.^{41,42,43,44}

On the other hand, QTMs still suffer from one major drawback—they are numerically highly unstable in the vicinity of nodes. This “node problem” manifests in several different ways:^{23,24} (1) infinite forces, giving rise to kinky, erratic trajectories; (2) compression/inflation of trajectories near wavefunction local extrema/nodes, leading to; (3) insufficient sampling for accurate derivative evaluations. In the best case, this can result in substantially more trajectories and time steps than the corresponding classical calculation; in the worst case, the calculation may fail altogether, beyond a certain point in

time.

For many molecular applications (though certainly not all), the initial wavepacket is nodeless; however, it may develop nodes at some later point in time. Moreover, from a practical standpoint, nodes per se are not the only source of numerical difficulty; in general, any large or rapid oscillations in the wavefunction—termed “quasinodes”²⁴—can be sufficient to cause the problems described above. Such oscillations are very easily formed in molecular systems, particularly during barrier reflection. Accordingly, several numerical techniques are being developed to address the node problem, including the Arbitrary Lagrangian-Eulerian frame method,^{45,46} and the artificial viscosity method.^{46,47}

In this paper, we take a different approach to the node problem, based on a thorough understanding of the differences and similarities between Bohmian and semiclassical mechanics. Formally, these two theories share many similarities, as was known from the earliest days^{13,48}—yet in practical terms, semiclassical and quantum trajectories often behave very differently. For instance, the former may cross in position space, but not in phase space; the latter do exactly the opposite. For the special case of stationary eigenstates in 1 DOF (the focus of the present paper), semiclassical trajectories evolve in phase space along the contours of the classical Hamiltonian, whereas quantum trajectories *do not move at all*. For well-behaved potentials, classical trajectories are always smooth and well-behaved, but quantum trajectories may be kinky and erratic.

As noted by Zhao and Makri,²⁸ nowhere are the differences between the two methods more profound than in the treatment of nodes and interference phenomena—which is not even *qualitatively* similar. In the semiclassical treatment, the approximate wavefunction is expressed in terms of simple functions that are generally as smooth and well-behaved as the potential itself. Oscillations and nodes are obtained when different lobes or “sheets” of the semiclassical wavefunction come to occupy the same region of space—thus giving rise naturally to interference. In contrast, the Bohmian representation of the wavefunction is single-valued, and therefore incapable of self-interference. Consequently, all of the undesirable—even “unphysical”—aspects of quantum trajectories, as described in the previous paragraph, are necessary in order to represent nodes and quasinodes explicitly.

From a dynamical perspective, the only difference between semiclassical and quantum trajectories is the quantum potential, Q , and it is a remarkable fact that Q alone is responsible for all of the very fundamental differences described above. On the other hand, this situation is also cause for alarm, for Q turns out to be the order \hbar^2 correction that is ignored in semiclassical treatments—being regarded as “insignificant” in the large action limit, in accord with the correspondence principle. From the previous discussion, it is clearly incorrect to regard Q as insignificant, which seems to place the semiclassical approximation—and indeed, the

correspondence principle itself—in jeopardy. Moreover, there are certain unappealing aspects of the semiclassical approach—multivaluedness, caustics, phase discontinuities, etc.^{13,15,16,17,19}—that simply do not arise in a Bohmian treatment. On the other hand, the semiclassical approximation is known to be valid in the large action limit—which together with the undesirable features of the Bohmian approach as discussed in the previous paragraph, seems to call into question the physical correctness of the latter. This paradox has been a source of concern for some researchers, notably Einstein.^{49,50,51}

The primary goal of the present paper is to reconcile the semiclassical and Bohmian theories, in a manner that preserves the best features of both, and also satisfies the correspondence principle. At least within the context of stationary eigenstates in 1 DOF, the above paradox turns out to be remarkably easy to resolve. It can be shown that the disturbing dissimilarities described above stem not from the theoretical methodologies themselves, but from the simple fact that each method uses a different ansatz to represent the wavefunction—thus giving rise to a rather unfair comparison. In particular, since the semiclassical functions are double-valued in the classically allowed region of space, the stationary wavefunction is represented as the sum of two terms—essentially a pair of “traveling waves,” headed in opposite directions. In contrast, the standard Bohmian approach uses a single term to represent the wavefunction. Virtually all of the disparities described above arise from this simple fact.

It is therefore natural to consider what would happen if the Bohmian formalism were applied to a *two-term* wavefunction—thus placing it on a proper par with the semiclassical method. As will be shown in this paper, this results in everything “falling into place.” In particular, the quantum potential—far from being singular in the vicinity of nodes—is well-behaved everywhere, and in fact, becomes vanishingly small in the large action limit, exactly in accord with the correspondence principle. The same can be said for the quantum trajectories, which are no longer stationary, and approach the corresponding semiclassical trajectories in the large action limit (within the classically allowed region of space). This implies the somewhat counterintuitive result that quantum trajectories must be well-behaved when the number of nodes is *large*, for this signifies the large-action limit. In any event, the two-term Bohmian approach provides us with the “best of both worlds,” i.e. the well-behaved trajectories of semiclassical mechanics, together with the singlevaluedness and lack of caustics and phase discontinuities that characterize Bohmian mechanics. More generally than for just the stationary states in 1 DOF considered here, it is anticipated that a multi-term Bohmian implementation will go a long way towards alleviating the node problem.

II. BACKGROUND

A. Unipolar ansatz

Let $\Psi(x)$ be any normalized wavefunction in the single DOF, x . Being a complex function, $\Psi(x)$ can be decomposed into two real functions, $R_B(x)$ and $S_B(x)/\hbar$, representing the amplitude and phase, respectively, as follows:

$$\Psi(x) = R_B(x)e^{iS_B(x)/\hbar} \quad (1)$$

Equation (1) is the celebrated Madelung-Bohm ansatz,^{12,25,26} which we term the “unipolar ansatz,”²⁴ as it consists of a single term only. The function $S_B(x)$ has units and interpretation of action. If time evolution is considered, then $S_B(x)$ plays the role of Hamilton’s principle function in classical mechanics,⁵² which therefore properly depends on t as well as on x . However, for stationary states in a time-independent context, $S_B(x)$ is analogous to Hamilton’s characteristic function, $W(x)$, which is time-independent. Both interpretations will be found to be important for the present approach.

The above decomposition is essentially unique if $R_B(x)$ is nonnegative throughout; however, it leads to discontinuities in $S_B(x)$, and cusps in $R_B(x)$, if $\Psi(x)$ has nodes. Despite these drawbacks, Eq. (1) is the decomposition generally utilized in standard Bohmian treatments,^{24,25,26} thus motivating use of the “ B ” subscript. It will be shown in this paper—evidently for the first time—that this convention in and of itself gives rise to certain node-related numerical difficulties that would otherwise not arise (Sec. II C).

Accordingly, for the present work, we presume amplitude functions that change sign when passing through nodes. To avoid confusion with the standard Bohmian decomposition, we use unsubscripted quantities to represent the present unipolar ansatz decomposition, i.e.

$$\Psi(x) = R(x)e^{iS(x)/\hbar}, \quad (2)$$

where $R_B(x) = |R(x)|$, $S_B(x) = S(x) \bmod (\pi\hbar)$, and the new decomposition functions, $R(x)$ and $S(x)$, are smooth and continuous throughout the entire coordinate range; for the latter reason, $S(x)$ is deemed a better analog for Hamilton’s functions than is $S_B(x)$. In any event, throughout this work, when discussing the unipolar ansatz, we are referring to Eq. (2), unless explicitly stated otherwise. Although the function $S(x)$ is unique modulo $2\pi\hbar$, physically, it is well-defined only up to the addition of an arbitrary constant, which introduces an arbitrary but immaterial phase factor into $\Psi(x)$.

Equation (2) is the starting point of both quantum trajectory *and* semiclassical methods. Presuming a quantum Hamiltonian of the form

$$\hat{H} = \frac{\hat{p}^2}{2m} + V(\hat{x}), \quad (3)$$

the general time evolution equations for the Eq. (2) unipolar decomposition functions are

$$\dot{R} = \frac{-1}{2m} (2R'S' + RS'') \quad (4)$$

$$\dot{S} = -V + \frac{1}{2m} \left[\hbar^2 \left(\frac{R''}{R} \right) - S'^2 \right], \quad (5)$$

where $R' = d[R(x)]/dx$, $\dot{R} = d[R(x)]/dt$, etc., and the coordinate dependences have been suppressed to save space. Equation (4) is the continuity equation, essentially stating that probability is conserved. Note that this equation is independent of the particular system potential, $V(x)$.

Equation (5) is the quantum analog of the Hamilton-Jacobi equation,⁵² which does depend on the particular $V(x)$. It is in the treatment of this equation that the semiclassical and Bohmian theories part company. The former ignores the first term in the square brackets, giving rise to the standard classical Hamilton-Jacobi equation. The latter regards the first term as the “quantum potential,”

$$Q(x) = -\frac{\hbar^2}{2m} \left(\frac{R''}{R} \right), \quad (6)$$

which is added to the true potential, to form the modified potential, $U(x)$. Apart from the substitution $V(x) \rightarrow U(x)$, the dynamical laws for the two approaches are identical.

In particular, in both cases, the momentum $P(x)$ is related to the action via

$$P(x) = S'(x) \quad (7)$$

The set of points $\{x, P(x)\}$ constitute a one-dimensional subspace of the 1 DOF phase space known as a “Lagrangian manifold” (LM).^{14,15,16,18} This terminology is generally used in a classical or semiclassical context only; however, we find it convenient to apply it in the exact quantum case as well.

B. Stationary states, and the bipolar ansatz

If $\Psi(x)$ is presumed to be a stationary state—i.e., an eigenstate of the Hamiltonian, \hat{H} , with energy E —then $\dot{R} = 0$, and $\dot{S} = -E$. The first result, together with Eq. (4), is consistent with the well-known property that for standing waves, the phase is constant over x . Without loss of generality, we may take $\Psi(x)$ to be real, so that $S(x) = 0$ and $R(x) = \Psi(x)$. The second result, together with Eq. (5), yields the Quantum Stationary Hamilton-Jacobi Equation (QSHJE). By rearranging Eq. (5), and making use of Eqs. (6) and (7), we obtain:

$$\begin{aligned} P^2(x) &= 2m[E - V(x) - Q(x)] \\ &= 2m[E - U(x)] \end{aligned} \quad (8)$$

An important connection between the semiclassical and Bohmian theories is suggested by Eq. (8)—namely, *the semiclassical approximation is accurate when the quantum potential is small*.

Let us now consider the semiclassical approximation proper—i.e., Eq. (8) with no $Q(x)$ term. The resultant algebraic equation has two solutions for $p(x)$, i.e. $p(x) = \pm\sqrt{2m[E - V(x)]}$ —where lower case ‘ p ’ is now used, for reasons that will be explained shortly. These two solutions correspond to the positive and negative momentum “sheets” of the LM in phase space. The two sheets are joined together at the classical turning points, x_{\min} and x_{\max} , to form a single LM in phase space, along the classical Hamiltonian contour, $H(x, p) = p^2/2m + V(x) = E$. (Turning points are the caustics for stationary states). Thus, the function $p(x)$ is double-valued over the classically allowed region, $x_{\min} \leq x \leq x_{\max}$, and zero-valued everywhere else.

It is illuminating to compare the semiclassical situation, as described above, with the Bohm prescription. From the correspondence principle, one expects $Q(x)$ to be small in the large action limit—i.e., the limit in which the excitation numbers, n , become large. This in turn would imply, via Eq. (8), that the semiclassical and exact quantum LMs would resemble each other in the large action limit. The actual situation is completely different, however. Firstly, Eqs. (2) and (7) imply that the quantum $P(x)$ is single-valued everywhere—rather than zero- or double-valued, like the semiclassical $p(x)$. Secondly, for stationary states, $S(x) = P(x) = 0$, implying that the quantum LM is the real axis of the phase space plane, which in no way resembles the semiclassical, Hamiltonian-contour LM. Thirdly, quantum trajectories are stationary over time, whereas semiclassical trajectories evolve around their LMs (Sec. III D).

The origin of these seemingly profound qualitative differences is deceptively simple: it is due to the fact that the semiclassical approximation does not actually incorporate the unipolar ansatz of Eq. (2). Instead, a *bipolar* ansatz is used for the total wavefunction, consisting of two terms rather than one:

$$\begin{aligned}\Psi(x) &= e^{i\delta} r(x) e^{is(x)/\hbar} + e^{-i\delta} r(x) e^{-is(x)/\hbar} \\ &= \Psi_+(x) + \Psi_-(x)\end{aligned}\quad (9)$$

In Eq. (9) above, the constant δ specifies the relative phase between the two components, $\Psi_{\pm}(x)$. It has been singled out from $s(x)$ in order to clarify certain issues pertaining to square-integrability (Sec. III B 2). Although $\Psi(x)$ itself is real, the $\Psi_{\pm}(x)$ are both complex, and conjugate to each other. The stationary “standing wave” is therefore obtained in practice as the linear superposition of two “traveling waves,” moving in opposite directions.

The standard Bohmian prescription, being unipolar, completely misses out on this elegant and useful aspect of the semiclassical approach, which gives rise to a qualitatively very different kind of amplitude/action decomposition. On the other hand, if the bipolar ansatz of Eq. (9) is incorporated into the Bohmian theory, rather

than Eq. (2), then it is indeed possible to reconcile these two approaches, in a manner consistent with the correspondence principle, as will be shown in Sec. III. Note, however, that an important difference between semiclassical and bipolar quantum LMs is already evident in Eq. (9); namely that the bipolar momentum function $p(x) = \pm s'(x)$ is double-valued throughout the entire coordinate range. Thus, the two exact quantum LM sheets never join, but extend into the classically forbidden regions all the way to $x = \pm\infty$.

Throughout this paper, we use lower case to denote the bipolar ansatz functions, so as to distinguish these from the unipolar ansatz functions, for which upper case is used. For convenience, all bipolar functions are hereafter defined to be single-valued everywhere, by referring to the positive-momentum LM sheet only—e.g., $p(x) = s'(x)$.

C. Node issues

The increased flexibility of the bipolar ansatz is extremely useful vis-a-vis the treatment of nodes, for it allows for the direct representation of nodes as *interference fringes arising naturally between the two traveling waves*. This possibility is exploited to great effect in semiclassical methods, which manage to contrive (approximate) bipolar amplitude functions $r(x)$ that are completely *nodeless*—no matter how many nodes are present in $\Psi(x)$ itself. Thus, apart from discontinuities near turning points (associated with Maslov indices^{14,15,17,18}), the semiclassical $r(x)$ is smooth and positive, and the semiclassical $s(x)$ is smooth and monotonically increasing. Moreover, these decomposition functions tend to be very slowly varying, in relation to $\Psi(x)$ itself, particularly when the latter has many nodes.

The above properties would of course also be beneficial for exact QTMs—which from a practical standpoint, is a primary reason why the bipolar ansatz ought to be considered within a Bohmian context. One difficulty is that the exact quantum bipolar decomposition of Eq. (9) is *not unique*, in the sense that the QSHJE of Eq. (8) has a two-parameter family of solutions.^{50,51} In particular, one trivial solution is $r(x) = \Psi(x)$; $s(x) = 0 = \delta$, which simply reproduces the unipolar result. Clearly, this is not the solution that we want, i.e., one that exhibits semiclassical correspondence in the large action limit; obtaining the latter will be the focus of Sec. III.

For the unipolar ansatz, it has been stated that nodes always give rise to infinities in $Q(x)$, owing to the singular denominator in Eq. (6). However, this is only strictly true if the standard Bohmian ansatz of Eq. (1) is used, for which the $R_B(x) \geq 0$ convention is employed. If instead, one adopts the Eq. (2) convention, so that $R(x)$ smoothly changes sign as a node is traversed, then Eq. (6) need not always be singular at a node.

In particular, $Q(x)$ is *never* singular when $\Psi(x)$ is a stationary eigenstate of \hat{H} , provided $V(x)$ is well-behaved everywhere. This is because the time-independent

Schrödinger equation guarantees that the nodes of $\Psi(x)$ also happen to be inflection points. Using Eq. (6) moreover, it can be shown that $Q(x) = E - V(x)$. Thus, for the stationary states considered in this paper, even the unipolar ansatz is not singular, contrary to what previously has been widely considered to be the case. Even if the standard Bohmian ansatz is used, $Q_B(x)$ at a node does not exhibit a singularity per se, but is rather ill-defined, owing to the cusp in $R_B(x)$; away from the nodal point, $Q_B(x) = Q(x)$.

In any event, we find it useful and convenient to distinguish between two types of nodes, depending on whether $Q(x)$ is formally well-behaved (“type one” nodes) or singular (“type two” nodes). From a numerical perspective, even type one nodes will cause difficulties for standard quantum trajectory calculations performed using the unipolar Bohmian ansatz. This is because the slightest numerical error in the evaluation of the Eq. (6) ratio will result in instability near the nodes—even though formally, $Q(x)$ does not diverge. In contrast, due to the smoothness and lack of nodes of the $r(x)$ functions that arise in the bipolar decomposition, numerical evaluation of the corresponding bipolar quantum potentials, $q(x)$, causes no such instabilities for type one nodes. More general nodal implications of the bipolar ansatz will be discussed in greater detail in future publications.

III. BIPOLAR DECOMPOSITION FOR STATIONARY STATES

A. Semiclassical properties

In this section, we derive a unique bipolar decomposition of the Eq. (9) form, for any given stationary wavepacket, $\Psi(x)$, which satisfies semiclassical correspondence in the large action limit. In general, the Eq. (9) decomposition is nonunique. The semiclassical solution, however, is essentially unique (Sec. IIB). We will therefore use the latter as a guide, for selecting the particular quantum bipolar decomposition which most closely resembles the semiclassical solution.

Note that certain assumptions have already entered into the form of Eq. (9), which is clearly more constrained than a completely general bipolar decomposition of $\Psi(x)$ into two arbitrary components. In particular, we have presumed $\Psi(x)$ to be a superposition of equal and opposite traveling waves—a natural assumption, completely analogous to the semiclassical situation. Be that as it may, there is still an enormous number of ways in which Eq. (9) may be realized for a given real wavepacket $\Psi(x)$, and so the decomposition is still far from being unique.

To help narrow the field, we first summarize some of the additional properties of semiclassical eigenstates in 1 DOF, which we will attempt to emulate in the exact quantum decomposition:

1. The LM itself *does not change* over time.

2. The classical probability distribution *does not change* over time.
3. The classical flow for either LM sheet maintains *invariant flux* over all x , with the flux value for the two sheets being equal and opposite.
4. The area enclosed within the LM, i.e. the enclosed action, J , is given by $J = 2\pi\hbar(n + 1/2)$, where n is the number of nodes.
5. For a normalized distribution, the absolute value of the invariant flux equals the inverse of the period of the trajectory, i.e. $F = \omega/2\pi$.
6. The median of the enclosed action, x_0 , satisfies $\int_{x_{\min}}^{x_0} p(x)dx = \int_{x_0}^{x_{\max}} p(x)dx$.
7. All trajectories move *along* the LM.

By “translating” these properties appropriately into the exact quantum context, we will be able to define an essentially unique bipolar decomposition of the Eq. (9) form.

B. Basic properties: (1)–(4)

1. invariant flux

Properties (1) and (2) above are the most fundamental, and will be considered first. For a particular Eq. (9) decomposition, the corresponding positive-momentum LM sheet is given by $p(x) = s'(x)$. Property (1) states that $\dot{p}(x) = 0$, which in turn implies $\dot{s}(x) = \text{const}$. Property (2) implies $\dot{r}(x) = 0$. Together, these properties imply that the $\Psi_{\pm}(x)$ components of $\Psi(x)$ must be stationary eigenstates of \hat{H} in their own right. Equation (9) then implies that the eigenvalues for the two components must both be equal to E .

Since the $\Psi_{\pm}(x)$ are stationary, the quantum mechanical flux associated with each of these components, i.e.

$$\begin{aligned} j_{\pm}(x) &= \frac{\hbar}{2im} \left[\Psi_{\pm}^*(x) \frac{d\Psi_{\pm}(x)}{dx} - \frac{d\Psi_{\pm}^*(x)}{dx} \Psi_{\pm}(x) \right] \\ &= \pm \left[\frac{p(x)}{m} \right] r^2(x), \end{aligned} \quad (10)$$

is independent of x , with equal and opposite constant values, $\pm F$. Note that Eq. (10) demonstrates that the quantum analog of property (3) is also satisfied—a necessary consequence of properties (1) and (2). We call this the “invariant flux” property.

For $F = 0$, $\Psi_+(x) = \Psi_-(x) \propto \Psi(x)$, which reproduces the unipolar ansatz. We shall therefore hereafter restrict consideration to the $F > 0$ case, for which the invariant flux property, and Eq. (10), provide a specification for r , in terms of s' , and the constant, F :

$$r(x) = \sqrt{\frac{mF}{s'(x)}} \quad (11)$$

Note that if $r(x) > 0$ for all x , then Eq. (11) implies that $p(x) > 0$ for all x —a desirable property for the positive momentum solution, also satisfied by the semiclassical solution. This would also imply that $s(x)$ is monotonically increasing. Accordingly, the $r(x) > 0$ condition is adopted.

For $F > 0$, the two $\Psi_{\pm}(x)$ components are linearly independent. This implies that at least one of the two must be non- L^2 . In fact, being complex conjugates of each other, *both* solutions must be non- L^2 . Moreover, it can be shown that $\Psi_{\pm}(x)$ diverges as $x \rightarrow \pm\infty$.^{53,54} This is due to the fact that $p(x) = s'(x) \rightarrow 0$ as $x \rightarrow \pm\infty$ in order that the enclosed action be finite (Sec. III B 2); but this implies via Eq. (11) that $r(x)$ diverges.

It is instructive to rederive the invariant flux property in another manner. By the superposition principle, the time evolution of $\Psi(x)$ can be obtained by propagating each of the $\Psi_{\pm}(x)$ components separately in time. Since the two components are stationary eigenstates of \hat{H} in their own right, the time-evolving $\Psi_{\pm}(x)$'s must each independently satisfy $\dot{r} = 0$ and $\dot{s} = -E$. The former, applied to a lower case version of Eq. (4), is equivalent to the spatial derivative of Eq. (10).

2. quantization

We now address the exact quantum analog of property (4), the quantization condition. In a proper semiclassical treatment, this half-integer condition on the enclosed action,^{55,56,57} $J = 2\pi\hbar(n + 1/2)$, must be supplemented by the discontinuous jumps in phase that occur as one traverses a turning point, from one LM sheet to another. In a certain sense, these jumps account for the fact that the WKB solutions do not incorporate the portion of the true wavefunction that tunnels into the forbidden region—which contribute an additional one half quanta of action, over the course of one complete circuit around the LM.¹⁹ When this discontinuous contribution is properly added to the usual enclosed action contribution, one obtains an *integer* quantization condition for the total action, $J_{\text{tot}} = 2\pi\hbar(n+1)$, even within a purely semiclassical context.

In the quantum case, there is no distinction between classically allowed and forbidden regions; one travels smoothly from one to the other, over the entire position space. Since the two LM sheets are symmetrically placed in phase space about the real axis (Fig. 1), the area enclosed between them is clearly twice the change in the action function, $\Delta s = \int_{x_{\min}}^{x_{\max}} p(x) dx$, as one travels from $x_{\min} = -\infty$ to $x_{\max} = +\infty$. From the above description, we expect this change in action to be $\pi\hbar(n+1)$, where n is the number of nodes. An integer quantization condition is therefore expected to hold for the exact bipolar quantum decomposition. This is indeed correct, as has been shown previously.^{53,54,58}

A sufficient, though certainly not necessary (see below), condition for achieving integer quantization of the

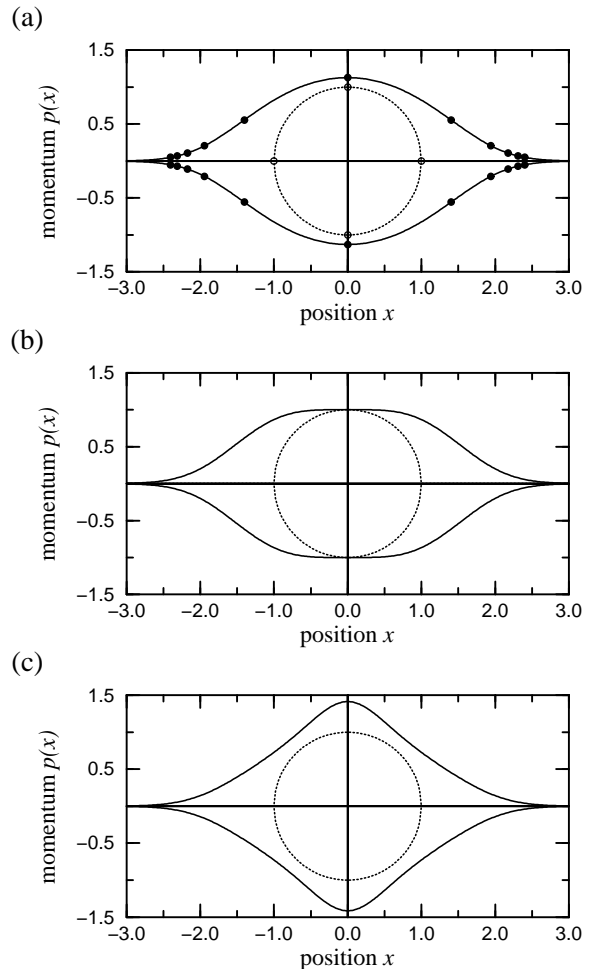


FIG. 1: Bipolar Lagrangian manifolds (LMs) for the harmonic oscillator ground state, $\Psi(x) = \exp(-x^2/2)/\pi^{1/4}$, for three different flux values, F ($x_0 = 0$ in each case): (a) $F = 1/2\pi \approx 0.159$, the semiclassical value; (b) $F = 1/4\sqrt{\pi} \approx 0.141$; (c) $F = 1/2\sqrt{2}\pi \approx 0.199$. Solid curves indicate quantum LMs; dotted curves indicate semiclassical LMs. The former enclose an area $J = 2\pi\hbar$; the latter $J = \pi\hbar$. In (a), the small open/filled circles represent semiclassical/quantum trajectory locations at times $t = k\pi/2$, for integer k .

quantum action is that $\Psi(x)$ be L^2 , which requires that $\Psi(\pm\infty) = 0$. For convenience, we adopt the convention that $s(-\infty) = -\pi\hbar(n+1)/2$ in Eq. (9). The condition $\Psi(-\infty) = 0$ then determines a value for δ —i.e., $\delta = 0$ for n even, and $\delta = \pi/2$ for n odd. This yields the following:

$$\Psi(x) = \begin{cases} 2r(x) \cos[s(x)/\hbar] & \text{for } n \text{ even;} \\ 2r(x) \sin[s(x)/\hbar] & \text{for } n \text{ odd.} \end{cases} \quad (12)$$

The somewhat awkward distinction between the even and odd n cases is due to our choice of boundary condition for $s(x)$; the reasons for this seemingly perverse choice will be made clear shortly.

Since $\Psi(+\infty)$ must also be zero if $\Psi(x)$ is L^2 , Eq. (12) implies that $s(+\infty) = \pi\hbar(n'+1)/2$, where n' is an integer. Since $r(x) > 0$ everywhere, all nodes of $\Psi(x)$, for n

even/odd, must occur at x values for which $s(x)/\hbar$ is an odd/even multiple of $\pi/2$. This, in turn, requires $n' = n$, from which is obtained

$$J = 2\Delta s = 2s(+\infty) - 2s(-\infty) = 2\pi\hbar(n+1), \quad (13)$$

i.e., the integer quantization condition. Note that $s(x)$ is independent of the normalization of $\Psi(x)$. However, the amplitude function $r(x)$ is not; thus, if $\Psi(x)$ is presumed normalized to unity, then Eq. (12) defines the normalization convention for $r(x)$, and for $\Psi_{\pm}(x)$.

The specific value of the constant, δ —as given above for even/odd n —is not arbitrary, but is required in order to ensure that the resultant $\Psi(x)$ be L^2 . If a different value were chosen, then a different, *non- L^2* solution to the Schrödinger equation would be obtained. This situation is in stark contrast to the unipolar case (for which the constant simply changes the overall phase), and is due to the fact that the bipolar δ represents a *relative* phase shift. In any event, we can regard δ as a parameter that is used to specify a particular solution of the Schrödinger equation at the energy E . In fact, *all* such real-valued solutions (apart from an immaterial scaling factor) may be obtained by varying the value of δ in Eq. (9). This is due to the fact that the $\Psi_{\pm}(x)$ are linearly independent. In any event, an important consequence is that all Schrödinger solutions are described by the exact same action function, $s(x)$, which is unaffected by the value of δ . Among other things, this implies that the integer quantization rule applies to all *non- L^2* solutions, as well as to the L^2 solution.

C. QSHJE properties: (5) and (6)

1. introduction

In this section, we continue the approach introduced at the end of Sec. III B 1, where the time evolution equations are applied to the two $\Psi_{\pm}(x)$ components separately. The $\dot{r} = 0$ equation was seen to yield the same invariant flux relation, for each $\Psi_{\pm}(x)$ component—i.e., Eq. (11). The $\dot{s} = -E$ equation, as applied separately to the two components, also yields identical results. One of the important ramifications of this is that the Eq. (9) ansatz is preserved over time, at least for stationary eigenstates.

In any event, since $\Psi_{\pm}(x)$ are solutions to the Schrödinger equation, the $\dot{s} = E$ equation must result in a lower-case version of the QSHJE [Eq. (8)], for which the bipolar quantum potential, $q(x)$, is defined via lower-case Eq. (6). We shall rewrite this QSHJE by expressing $q(x)$ directly in terms of $p(x)$, using lower-case Eqs. (6) and (7), and Eq. (11):⁵⁹

$$p^2 = 2m(E - V) - \hbar^2 \left[\frac{1}{2} \left(\frac{p''}{p} \right) - \frac{3}{4} \left(\frac{p'}{p} \right)^2 \right] \quad (14)$$

Whereas the semiclassical approximation [obtained by ignoring the $q(x)$ term in brackets] has a unique $p(x) > 0$

solution, Eq. (14) is a second-order differential equation in $p(x)$, with a two-parameter *family* of different solutions to choose from.

Note that Eq. (14) applies to *all* Schrödinger equation solutions, i.e. the L^2 and the non- L^2 solutions, both. Since we are interested only in the former, and since there is a one-parameter family of Schrödinger solutions in all, one might expect that the specification of the L^2 solution would determine the value of one of the two parameters. This is not correct however, as demonstrated earlier (Sec. III B 2). Thus, even for the L^2 solution alone, bipolar decomposition gives rise to two variable parameters via Eq. (14).

2. defining the two parameters

What are the two parameters, and how should their values be chosen? To determine what the two parameters are, it is convenient to combine Eqs. (11) and (12) together, to obtain a formula for $\Psi(x)$ directly in terms of $s(x)$. The result is:

$$\Psi(x) = \begin{cases} \sqrt{4mF/s'(x)} \cos[s(x)/\hbar] & \text{for } n \text{ even;} \\ \sqrt{4mF/s'(x)} \sin[s(x)/\hbar] & \text{for } n \text{ odd.} \end{cases} \quad (15)$$

Equation (15) is a first-order differential equation for $s(x)$; the general solution is easily found to be

$$\begin{aligned} \tan(s/\hbar) &= \frac{4mF}{\hbar} \int_{x_0}^x \frac{dx'}{\Psi^2(x')} & \text{for } n \text{ even;} \\ -\cot(s/\hbar) &= \frac{4mF}{\hbar} \int_{x_0}^x \frac{dx'}{\Psi^2(x')} & \text{for } n \text{ odd.} \end{aligned} \quad (16)$$

For nodeless wavepackets ($n = 0$), $\Psi(x) > 0$ everywhere, and the integrand of Eq. (16) has no singularities. When $n > 0$, Eq. (16) is still correct, but requires careful branch selection, to ensure that the final $s(x)$ curve is continuous throughout the coordinate range. Note that $\Psi(x)$ is presumed to be the L^2 Schrödinger solution.

Equation (16) provides an explicit recipe for obtaining the Eq. (12) decomposition. The two parameters can thus be taken as: (1) the flux parameter, F ; (2) the integration limit parameter, x_0 . Note that $s(x_0) = 0$; consequently, x_0 may also be interpreted as the median of the action, as per Sec. III A. By varying the two parameters F and x_0 in Eq. (16), different bipolar decompositions may be achieved. These correspond to different *affine transformations* of each other, in the sense that varying F is equivalent to *rescaling* the right-hand-side (RHS) of Eq. (16), whereas varying x_0 is equivalent to *adding a constant* to the RHS.

3. choosing parameter values

For a given $\Psi(x)$, the various bipolar LMs that can be constructed via Eq. (16) vary significantly with respect

to F and x_0 (Sec. IV), and so a general procedure for obtaining reasonable parameter values must be provided. At present, the best approach seems to be to touch base once again with the semiclassical properties—in particular, property (5) for determining the appropriate value of F , and property (6) for determining the appropriate value of x_0 .

Semiclassically, the flux for a normalized distribution is given by $F = \omega/2\pi$, where ω is the classical angular frequency for the appropriate semiclassical trajectory—i.e., the (uniform) rate at which the angle variable of the action/angle pair changes. The corresponding quantum trajectory is not that of a bound state, and so it is not possible to assign an ω value to it (Sec. III D). On the other hand, the Eq. (12) normalization convention allows us to determine a unique flux value for the quantum trajectory, which is all that is required. By setting the quantum flux value equal to the semiclassical value, it is anticipated that the resultant quantum LMs will closely resemble the semiclassical LMs, as desired.

As for x_0 , the median of the enclosed action: this can be regarded as the exact middle of the wavepacket in a certain sense; semiclassically, x_0 is the classically allowed configuration that is furthest from both of the turning points, vis-a-vis the action. Consequently, we expect the greatest agreement of semiclassical and quantum LMs—i.e. the smallest $q(x)$ values—in the vicinity of the semiclassical x_0 . This can be achieved by allowing the quantum x_0 to coincide with the semiclassical value—i.e., the latter is chosen to be the location where $s = 0$.

In the quantum bipolar decomposition scheme—even for fixed F —one is otherwise free to place the action median, x_0 , essentially anywhere along the position axis. The ramifications are particularly illuminating when $V(x)$ is even. For such potentials, *only* the choice $x_0 = 0$ gives rise to quantum bipolar decomposition functions that are even or odd in x , thereby respecting the physical symmetry of the system, and of $\Psi(x)$ itself. This choice for x_0 is also consistent with the median action criterion. Presumably, it would be unphysical to consider any of the asymmetrical decompositions; nevertheless, it is interesting to note that one can generate *asymmetrical* bipolar decompositions that give rise to the *symmetrical* $\Psi(x)$, simply by shifting x_0 away from the origin. This has been verified via explicit construction for the harmonic oscillator ground state.

D. Quantum trajectories: property (7)

We now address the issue of the quantum trajectories themselves, related to property (7). Semiclassically, over the course of time, the bound state trajectories simply move around and around the Hamiltonian contour LMs, which do not themselves change [property (1)]. In a conventional unipolar quantum treatment, the initial LM—specifying the initial conditions for the

ensemble of trajectories—is just the real axis, i.e. the “curve” $P(x) = 0$, since $\Psi(x)$ is real. The quantum trajectories evolve under the unipolar modified potential, $U(x) = V(x) + Q(x)$, which by Eq. (6), must be the constant function $U(x) = E$ [even if there are nodes (Sec. II C)]. Consequently, $\dot{P} = -U'(x) = 0$, and so the unipolar quantum trajectories *do not move at all* over time.

In contrast, the bipolar quantum trajectories are *not* stationary, but move along the positive and negative momentum LM sheets. This is true because $p(x) > 0$, and because the bipolar LMs themselves do not change over time, thus verifying property (7). Moreover, provided the bipolar quantum potential $q(x)$ is small in the classically allowed region, then the bipolar quantum trajectories must resemble the semiclassical trajectories within this region, since the LMs are similar, and $\dot{p} = -u'(x) \approx -V'(x)$. Of course, the bipolar quantum trajectories *do not* change their direction at the classical turning points, moving between positive and negative-momentum LM sheets, like classical trajectories. Instead, all quantum trajectories on say, the positive momentum LM sheet, continue to head to the right for all time. Once these trajectories enter the classically forbidden region, however, their speed decreases very suddenly.

It is worth discussing the very different role played by the quantum potential in the unipolar ansatz, versus that of the bipolar ansatz with the specific decomposition suggested here (i.e., parameter choices of Sec. III C 3). In the unipolar case, $Q(x)$ serves to counteract the true potential everywhere; thus, $Q(x)$ is not generally small. In contrast, the bipolar quantum potential, $q(x)$, can be regarded as the $\mathcal{O}(\hbar^2)$ correction to the semiclassical approximation—in the truest correspondence-principle sense of lower-case Eq. (8). The value of $q(x)$ is therefore small in the appropriate semiclassical limits—i.e., in the classically allowed region far from turning points, and in the limit of large action, when n becomes large. Near the turning points, $|q(x)|$ increases substantially, so as to ensure that all trajectories keep moving past the classical turning point without changing direction. This increase continues well into the classically forbidden region, where curiously, $q(x)$ approaches $Q(x)$ —i.e. it effectively cancels out the true potential. Consequently, the bipolar modified potential, $u(x) = V(x) + q(x)$, resembles the true potential in the classically allowed region, and the unipolar modified potential, $U(x) = E$, in the asymptotic regions.

The above discussion hinges on the assumption that the semiclassical and bipolar quantum LMs become arbitrarily close in the appropriate semiclassical limits described above. We can justify this expectation as follows. First, the semiclassical approximation is known to become arbitrarily accurate in these limits; each of the two semiclassical traveling wave components must therefore approach some particular corresponding pair of exact quantum solutions, $\Psi_{\pm}(x)$, arbitrarily closely. The latter must therefore have the same characteristics, vis-

a-vis action, trajectories, and flux, as do the semiclassical approximations, in the appropriate limits. Therefore, by choosing the available parameters for the quantum solutions (i.e. F and x_0) so as to match the semiclassical approximations, the correspondence principle must be satisfied.

E. Stationary non-eigenstates

Although the primary interest of this paper is bound, stationary eigenstates of the Hamiltonian \hat{H} of Eq. (3), our ultimate interest is wavepackets that evolve dynamically over time. As a first step in this direction, we generalize the previous discussion to include wavepackets that are only momentarily “stationary.” In other words, the initial wavepacket $\Psi(x)$ is real, but otherwise arbitrary, i.e., not presumed to be an eigenstate of \hat{H} . This results in $\dot{r} = 0$, but only instantaneously, at the initial time. We shall call such a wavepacket a “stationary non-eigenstate.”

To what extent can the bipolar decomposition scheme be applied to stationary non-eigenstate wavepackets? The question is relevant, because it is only necessary to specify the bipolar decomposition at a single point in time, in order to propagate the two $\Psi_{\pm}(x)$ components independently, over all time. Our approach shall be to regard $\Psi(x)$ as the eigenstate of some Hermitian, Hamiltonian-like operator, \hat{H}_0 , which without loss of generality, may be taken to be of the form

$$\hat{H}_0 = \frac{\hat{p}^2}{2m} + V_0(\hat{x}). \quad (17)$$

If $\Psi(x)$ is known, it is a trivial matter to obtain $V_0(x)$ by solving the Schrödinger equation in reverse, i.e.

$$[V_0(x) - E_0] = \frac{\hbar^2}{2m} \left(\frac{\Psi''}{\Psi} \right). \quad (18)$$

The \hat{H}_0 so obtained can then be used to generate the appropriate F and x_0 values semiclassically.

As a completely general procedure, this approach has one unavoidable flaw. If $\Psi(x)$ has type two nodes, then $V_0(x)$ will have singularities at the nodes, which is undesirable. In such cases, since the precise values of the F and x_0 parameters may not matter all that much in numerical practice, one should simply choose “reasonable” values by comparison with known cases—e.g., for $n = 0$, one could use the parameters of a Gaussian with the same center and standard deviation as $\Psi(x)$. On the other hand, almost all initial wavepackets used in chemical physics applications correspond [via Eq. (18)] to potentials $V_0(x)$ that are well-behaved.

IV. RESULTS: HARMONIC OSCILLATOR EIGENSTATES

As a classic benchmark example, we now work out analytic solutions for the harmonic oscillator (HO) eigenstates, i.e. $V(x) = kx^2/2$. This example is particularly important, as the ground state provides the proper decomposition for Gaussian wavepackets, which are used very frequently in time-dependent studies. We shall also find the excited harmonic oscillator states to be quite useful, particular with respect to investigations regarding nodes and interference. The normalized n 'th harmonic oscillator eigenstate is given by

$$\Psi_n(x) = (2^n n!)^{1/2} \left(\frac{m\omega}{\hbar\pi} \right)^{1/4} \times H_n \left(\sqrt{\frac{m\omega}{\hbar}} x \right) e^{-m\omega x^2/2\hbar}, \quad (19)$$

where $H_n()$ is the n 'th Hermite polynomial, and $\omega = \sqrt{k/m}$.

A. Ground state

We start with the ground state, $\Psi_0(x) = (m\omega/\hbar\pi)^{1/4} e^{-m\omega x^2/2\hbar}$. Application of Eq. (16) yields

$$s(x) = \hbar \arctan \left\{ F \left(\frac{2\pi}{\omega} \right) \times \left[\operatorname{erfi} \left(\sqrt{\frac{m\omega}{\hbar}} x \right) - \operatorname{erfi} \left(\sqrt{\frac{m\omega}{\hbar}} x_0 \right) \right] \right\} \quad (20)$$

as the generic, F - and x_0 -dependent solution. This gives rise via Eq. (11) to

$$r(x) = \left(\frac{1}{2} \right) \left(\frac{m\omega}{\hbar\pi} \right)^{1/4} e^{-m\omega x^2/2\hbar} \times \left\{ 1 + F^2 \left(\frac{2\pi}{\omega} \right)^2 \times \left[\operatorname{erfi} \left(\sqrt{\frac{m\omega}{\hbar}} x \right) - \operatorname{erfi} \left(\sqrt{\frac{m\omega}{\hbar}} x_0 \right) \right]^2 \right\}^{1/2}. \quad (21)$$

As per Sec. III C 3, the appropriate value of F is clearly $F = \omega/2\pi$. The appropriate value of x_0 , whether from symmetry considerations, or the more general median action criterion, is clearly $x_0 = 0$. With these choices for the parameter values, we obtain the simpler result

$$s(x) = \hbar \arctan \left[\operatorname{erfi} \left(\sqrt{\frac{m\omega}{\hbar}} x \right) \right], \quad (22)$$

$$r(x) = \left(\frac{1}{2} \right) \left(\frac{m\omega}{\hbar\pi} \right)^{1/4} e^{-m\omega x^2/2\hbar} \times \left[1 + \operatorname{erfi}^2 \left(\sqrt{\frac{m\omega}{\hbar}} x \right) \right]^{1/2}. \quad (23)$$

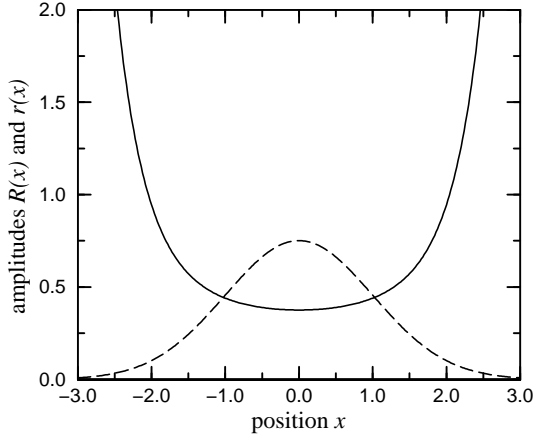


FIG. 2: Amplitude functions for the harmonic oscillator ground state, $\Psi(x) = \exp(-x^2/2)/\pi^{1/4}$. Dashed curve: unipolar amplitude, $R(x) = \Psi(x)$. Solid curve: bipolar amplitude, $r(x)$, for semiclassical parameter values, $F = 1/2\pi$ and $x_0 = 0$.

For convenience, we choose units such that $\hbar = m = k = \omega = 1$. In these units, $F = 1/2\pi$, and $x_0 = 0$. In these units, and for these parameter values, all of the relevant bipolar decomposition functions are as follows:

$$\begin{aligned} s(x) &= \arctan[\operatorname{erfi}(x)] \\ r(x) &= \left(\frac{1}{2}\right) \left(\frac{1}{\pi}\right)^{1/4} e^{-x^2/2} [1 + \operatorname{erfi}^2(x)]^{1/2} \\ p(x) &= \frac{2e^{x^2}}{\sqrt{\pi} [1 + \operatorname{erfi}^2(x)]} \\ q(x) &= \frac{1}{2} - \frac{x^2}{2} - \frac{2e^{2x^2}}{\pi [1 + \operatorname{erfi}^2(x)]^2} \end{aligned} \quad (24)$$

All of the functions in Eq. (24) are smooth, slowly varying, and monotonic in $|x|$. The LM is an elegant “eye-shaped” curve [specified by the $p(x)$ equation above] that deviates smoothly, and positively, from the circular semiclassical LM, with the point of closest approach being $x_0 = 0$. All of these features are as predicted in Sec. III, and would not have been satisfied if substantially different parameter values were used. A plot of the semiclassical and bipolar quantum LMs is presented in Fig. 1, for $F = 1/2\pi$ and other values (but all with $x_0 = 0$). Whereas some of these other plots have the qualitatively correct behavior, it is very clear that the $F = 1/2\pi$ curve is the smoothest, most “correct” choice—especially vis-à-vis comparison with the corresponding semiclassical LM.

Figure 2 is a comparison between the unipolar and bipolar amplitude functions—i.e., $R(x)$ and $r(x)$, respectively. As is clear from the figure, these two types of amplitude behave completely differently. In particular, whereas $R(x)$ decreases quickly as $x \rightarrow \pm\infty$, $r(x)$ increases as one moves away from the origin, and actually diverges in the $x \rightarrow \pm\infty$ limits, as predicted in Sec. III B 1. Clearly, the $\Psi_{\pm}(x)$ are non- L^2 solutions.

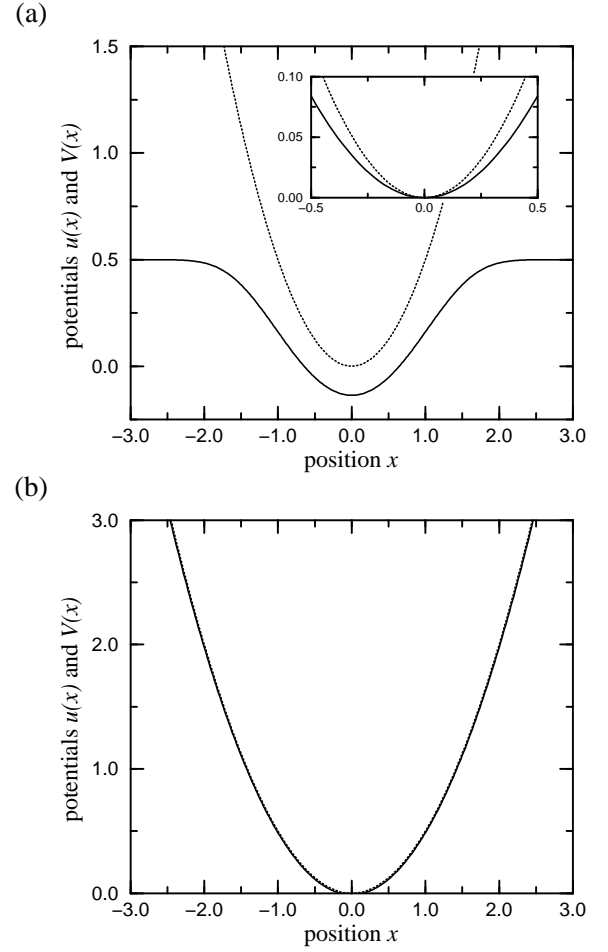


FIG. 3: Bipolar modified potentials, $u(x)$, and true potential, $V(x)$, for two different harmonic oscillator eigenstates: (a) ground state, $n = 0$; (b) tenth excited state, $n = 10$. Solid curves indicate $u(x)$; dotted curves indicate $V(x)$. In (a), $u(x)$ approaches $E_{n=0} = 1/2$ as $x \rightarrow \pm\infty$, but resembles $V(x)$ in the classically allowed region, $|x| < 1$ [inset shows $u(x) - u(0)$ vs. $V(x)$]. In (b), $u(x)$ and $V(x)$ are virtually indistinguishable over the (classically allowed) coordinate range indicated.

In Fig. 3(a), we present a comparison of the actual and bipolar modified potentials—i.e., $V(x)$, and $u(x) = V(x) + q(x)$. The two potentials resemble each other in the classically allowed region, away from the turning points at $x = \pm 1$. As one approaches the turning points, the difference $q(x)$ increases markedly. In the classically forbidden region, $u(x)$ ceases to emulate the true potential, and in the asymptotic limits, approaches the unipolar $U(x)$ constant value of $E = 1/2$. All of this is as predicted in Sec. III D.

We also performed trajectory calculations. In particular, a single trajectory was propagated over a very long period of time, using what Wyatt has called the “analytical approach.”²⁴ In this scheme, the modified force [i.e. $-u'(x)$] is computed analytically, but the trajectory itself is propagated numerically. We found first of all that this

numerical propagation scheme was extremely stable, as demonstrated by the fact that the numerical trajectory did not deviate appreciably from the LM at any point in time. Phase space values for the trajectory at various times are indicated as small circles in Fig. 1(a), from which it is also clear that quantum trajectories correspond fairly well to the semiclassical trajectories in the classically allowed region.

In the classically forbidden regions, trajectories slow down very quickly, as predicted. This is evidenced by the pile-up of trajectory points that ensues in these regions [Fig. 1(a)]. Formally, however, the trajectories do not actually reach zero momentum until $x \rightarrow \infty$. They are thus analogous to classical trajectories for a system that has just enough energy for dissociation. This fact is also reflected in the asymptotic behavior of $u(x)$ as discussed above.

We now briefly address the issue of trajectory “pile-up” in the classically forbidden regions, which is an important concern for numerical calculations. Although the bipolar ansatz has the effect of placing more trajectories in regions of space where the actual probability is small, this situation is numerically agreeable for two reasons: (1) more accuracy is needed in these regions, because $\Psi(x)$ itself is effectively obtained as the difference between two large numbers; (2) a simple “recycling” scheme can be introduced to reduce the number of trajectories to a minimum. These issues will be discussed in great detail in future publications.

B. Excited states

The correspondence between the semiclassical and quantum LMs for the harmonic oscillator ground state is only fairly good, but one ought to recall that the action is minimal in this case. A real test of the correspondence principle requires a detailed investigation of the LM behavior in the large action limit. This in turn, requires that the bipolar decomposition be performed for the excited harmonic oscillator states. Using Eq. (16), with $\hbar = m = k = \omega = 1$, $F = 1/2\pi$, and $x_0 = 0$, we have obtained analytical solutions for all n up to $n = 10$.

The general form of the bipolar action for the n th eigenstate [denoted $s_n(x)$] is as follows:

$$s_n(x) = \begin{cases} \arctan \left[\frac{e^{x^2} f_n(x)}{\sqrt{\pi} g_n(x)} + \operatorname{erfi}(x) \right] & \text{for } n \text{ even;} \\ -\operatorname{arccot} \left[\frac{e^{x^2} f_n(x)}{\sqrt{\pi} g_n(x)} + \operatorname{erfi}(x) \right] & \text{for } n \text{ odd.} \end{cases} \quad (25)$$

In Eq. (25) above, $f_n(x)$ is an $(n-1)$ th-order odd/even polynomial, and $g_n(x)$ is an n th-order even/odd polynomial, for n even/odd. Explicit coefficient values for $f_n(x)$ and $g_n(x)$ are listed in Tables I and II, respectively (coefficients for larger n can be provided on request).

The bipolar solutions for the excited states behave exactly as predicted. In particular $r_n(x) > 0$ everywhere,

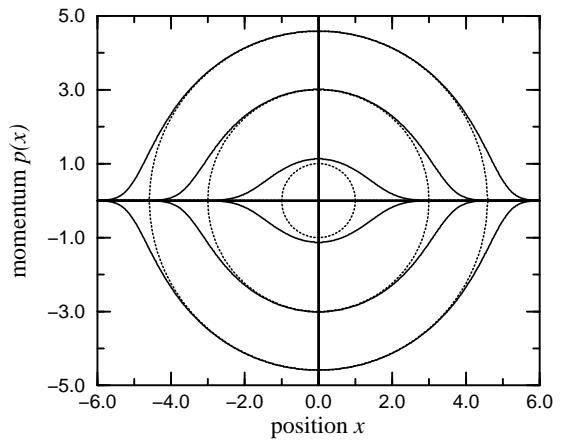


FIG. 4: Bipolar Lagrangian manifolds (LMs) for three different harmonic oscillator eigenstates; moving concentrically outward from the origin, these are $n = 0$, $n = 4$, and $n = 10$, respectively. Solid curves indicate quantum LMs; dotted curves indicate semiclassical LMs. The correspondence principle is clearly satisfied with increasing n .

and both $r_n(x)$ and $s_n(x)$ are as smooth, slowly varying, and monotonic as for the ground state. In fact, all of the bipolar functions qualitatively resemble those for $n = 0$, except on a larger scale—as is also true of the semiclassical functions. This is in sharp contrast to the behavior of the wavefunctions, $\Psi_n(x)$, themselves, which gain nodes and rapid oscillations as n is increased. One very encouraging aspect of the excited state bipolar functions is that nodal features are not evident anywhere. However, this requires choosing the correct branch of the Eq. (16) solution at every point in position space, such that the resultant $s_n(x)$ curve is not discontinuous across a node. This issue is discussed in more detail in Sec. IV C.

In any case, the basic goal of the bipolar ansatz has been achieved—i.e., to obtain a decomposition which, like the corresponding semiclassical approximation, treats all nodes as interference between a superposition of left and right traveling waves, $\Psi_{\pm}(x)$, which are themselves nodeless. Moreover, beyond achieving just this basic goal, we find that the *correspondence principle is satisfied in the large n limit*. This is exemplified in Fig. 4, wherein the semiclassical and bipolar quantum LMs are presented for several harmonic oscillator states over the n range considered.

From the figure, the quantum LMs are seen to enclose one-half quanta of area more than the semiclassical LMs, which manifests primarily in the forbidden regions near the turning points, as expected. In a relative sense, this discrepancy becomes decreasingly significant in the large n limit. Note that the quantum LMs completely enclose the semiclassical LMs, which was not anticipated earlier, but is certainly a desirable property. From lower-case Eq. (8), this will only be satisfied if the bipolar quantum potential is negative everywhere. This has been observed for all examples considered thus far, using the appropriate semiclassical values for F and x_0 ; however, most other

TABLE I: Coefficients, a_j , for polynomials, $f_n(x) = \sum_{j=0}^{n-1} a_j x^j$, for all $0 < n \leq 10$. These polynomials appear in the numerator of the analytic formula for the bipolar action, $s_n(x)$ [Eq. (25)].

Order	Coefficients									
n	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7	a_8	a_9
1	1									
2		2								
3	2		-2							
4		10		-4						
5	8		-18		4					
6		66		-56		8				
7	48		-174		80		-8			
8		558		-740		216		-16		
9	384		-1950		1380		-280		16	
10		5790		-10560		4704		-704		32

TABLE II: Coefficients, b_j , for polynomials, $g_n(x) = \sum_{j=0}^n b_j x^j$, for all $0 < n \leq 10$. These polynomials appear in the denominator of the analytic formula for the bipolar action, $s_n(x)$ [Eq. (25)].

Order	Coefficients										
n	b_0	b_1	b_2	b_3	b_4	b_5	b_6	b_7	b_8	b_9	b_{10}
1		-1									
2	1		-2								
3		-3		2							
4	3		-12		4						
5		-15		20		-4					
6	15		-90		60		-8				
7		-105		210		-84		8			
8	105		-840		840		-224		16		
9		-945		2520		-1512		288		-16	
10	945		-9450		12600		-5040		720		-32

parameter choices would not satisfy this property.

C. Numerical issues

Even more so than in the ground state case, the bipolar quantum potential for $\Psi_n(x)$ is found to be very small throughout most of the classically allowed region. The magnitude of $q_n(x)$ decreases with increasing n , so that whereas $q_0(0) = -0.137$ for the ground state, by $n = 10$ we have $q_{10}(0) = -0.012$. Of course, the extent of the classical region is also larger with increasing n ; thus by $n = 10$, we find $u_{10}(x)$ to be practically indistinguishable from the $V(x)$ over the range $|x| \leq 3$. The situation, depicted in Fig. 3(b), can be regarded as another manifestation of the correspondence principle.

The correspondence principle also has important ramifications for trajectory calculations. In particular, not only are the bipolar quantum trajectories for large n smooth and well-behaved throughout, but in the classically allowed region, they are virtually indistinguishable from classical trajectories. This has once again been verified by performing analytical trajectory calculations for $n = 10$, which were found to be just as numerically stable as for the ground state—despite the fact that $\Psi_{10}(x)$ itself has *ten nodes*. This bodes very well for obviating the node problem in general.

Although the bipolar decomposition functions—once obtained—exhibit no special behavior in the vicinity of nodes, it turns out that nodes complicate the determination of these functions somewhat, vis-a-vis implementation of Eq. (16). To begin with, let us imagine that—as in the current harmonic oscillator case—an analytical expression for the Eq. (16) integral is available. For the moment, we also take n to be even. Note that the left and right sides of Eq. (16) must be infinite at the nodes. Thus, whereas the exact analytical expression can be used across the entire coordinate range, a new branch is encountered each time a node is traversed. The specific branch of interest is specified by the condition of continuity for $s_n(x)$, and by $s_n(x_0) = 0$.

A superficial difficulty is encountered for the odd n states, for which there is necessarily a node at x_0 . Strictly speaking, this implies that the Eq. (16) integration must be singular. To circumvent this difficulty, we express the RHS of Eq. (16) as an indefinite analytical integral, plus an arbitrary constant, B . Note that since x_0 must lie at a node for odd n , x_0 can not serve as the second parameter, for singling out the particular solution of interest for Eq. (14). We can, however, use B for this

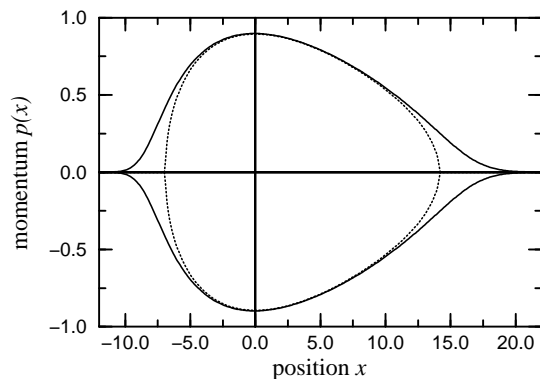


FIG. 5: Bipolar Lagrangian manifolds (LMs) for the fourth excited eigenstate of the Morse oscillator system with twenty bound states total. Solid curves indicate quantum LMs; dotted curves indicate semiclassical LMs. Semiclassical values for F and x_0 were used to specify the quantum solution, as per Sec. IV C.

purpose. In particular, if $V(x)$ is even, then only one value of B gives rise to the requisite odd $s_n(x)$ solution. More generally, i.e. for arbitrary $V(x)$, we can still apply the oddness criterion locally. In other words, it is easy to show that B should in general be chosen such that $s_n(x_0 - \epsilon) = -s_n(x_0 + \epsilon)$ in the limit of small ϵ . This technique bears a resemblance to the Cauchy principal value method.⁶⁰

The above discussion presumes that an analytical integral is available for the RHS of Eq. (16). Generally speaking, this will not be the case, and we must consider how to apply the above procedures when the integrations are performed numerically. Fortunately, this is straightforward. The general procedure is to pick an arbitrary integration limit, x_i , lying in between each adjacent pair of nodes (where $x = \pm\infty$ are treated as “nodes” in this context). Equation (16) is then applied to each interval separately, generating a smooth, numerically integrated function over the entire i ’th interval, that is correct to within an additive constant, B_i . The individual B_i values are then obtained, using the constraint $s_n(x_0) = 0$, and applying the Cauchy-like condition described above across each of the nodes separately.

The numerical procedure described above has been applied successfully to the Morse oscillator system, for which: (1) $V(x)$ is not symmetrical, and; (2) the Eq. (16) integrations must be performed numerically. The results will be presented in a future paper. We mention the Morse investigation in this paper simply to emphasize the fact the present method is in fact applicable in a much broader context than the analytical harmonic oscillator system considered here. Moreover, all of the conclusions drawn for the harmonic oscillator, regarding the correspondence principle and the like, evidently apply to more general systems. This is demonstrated in Fig. 5, which depicts the semiclassical and bipolar quantum LMs for the $n = 4$ state of the Morse oscillator.

V. COMPARISON WITH RELATED METHODS

The present work does not constitute the first application of the bipolar ansatz in a Bohmian-like dynamical context. For two decades or so, a very interesting bipolar approach has been developed and advocated by E. R. Floyd.^{50,51,54,61,62,63} More recently, essentially the same technique was derived by Faraggi and Matone (FM),^{51,64,65} within a much broader context, and using a very different physical picture. It is somewhat remarkable that these two approaches give rise to exactly the same dynamical equations (an illuminating comparison is presented in Ref. 24 and in Ref. 51). Perhaps even more remarkable, however, is that the dynamical law used in Floydian/FM trajectory propagation is *not* equivalent to that of Bohmian mechanics.

In this section, we compare and contrast the methods of Floyd, FM, and the present work. The various approaches were developed independently, and so a brief discussion of the different philosophies is presented, as well as the mathematical similarities and differences. The comparison is particularly apt for the present paper, in that both the Floyd and FM theories are restricted to *stationary states only*.

The starting point of the Floydian approach is the QSHJE in 1 DOF (multidimensional systems may be considered, but only if there is separation of variables⁵¹). As per Sec. II, this is natural enough, for a Bohmian-like theory applied to stationary states; however, there are some subtleties regarding the manner in which energy is treated, that give rise to a non-Bohmian dynamical law. The intriguing approach of FM begins not with the QSHJE, but with the fundamental postulate that all systems are equivalent under coordinate transformations.^{51,65} This is termed the “equivalence principle,” in obvious analogy with general relativity, with which the FM approach shares many parallels. Indeed, one of the goals of FM is to reconcile quantum mechanics and general relativity, to which end it is natural to focus on the action, which plays a key role in both physical theories.

In any event, the FM approach relies on the existence of a particular coordinate transformation that reduces the system to that of a free particle.^{51,64} This approach implicitly relies on the fact that the Wigner-Weyl correspondence is not preserved under canonical/unitary transformations⁵⁸—a feature that has been exploited to great effect in improvements to semiclassical theories, such as the Langer modification,^{66,67} although not always with full comprehension that this is what was taking place.^{68,69}

Starting from the equivalence principle, FM *derive* the QSHJE, and demonstrate that energy quantization arises naturally from the condition that this postulate be satisfied. They find, moreover, that a unipolar ansatz is insufficient to achieve this, but rather, a bipolar ansatz of the Eq. (9) form is required. In contrast, Floyd begins with the QSHJE, and then presumes a bipolar ansatz

for which each of the two terms is a solution. Floyd's motivations are evidently similar to the author's, in that he introduces the bipolar ansatz in order to:^{50,51} (1) obtain a Bohm-like formalism that is well-behaved at nodes; (2) obtain dynamical trajectories that are *not* stationary, thus obviating Einstein's concern, and allowing for the possibility of satisfying the correspondence principle.

For purposes of this discussion, the approaches of Floyd and of FM may from this point forward be regarded as identical. Since both employ the QSHJE, as does the present approach, it is clear that *all three utilize essentially the same bipolar functions*, $s(x)$, $r(x)$, $p(x)$, and $q(x)$. However, there are some important differences. Floyd uses a different convention, for which both the normalization of $\Psi(x)$, and the flux, change simultaneously, so that his $r(x)$ is proportional to ours. Neither of the other methods identifies the two parameters associated with Eq. (14) in the way that we have done, and they certainly do not provide a means of selecting preferred values for these parameters, as per Sec. III C 3.

Indeed, Floyd considers each of the two-parameter family of solutions to Eq. (14), which he terms “microstates,” to constitute an equally valid decomposition of $\Psi(x)$. He further asserts that since the Schrödinger equation per se provides no means of distinguishing microstates, that the QSHJE must be regarded as the more fundamental equation, in some sense. Floyd also acknowledges the fundamental differences between $q(x)$ and $Q(x)$, and the advantages of the former vis-a-vis nodes, as discussed in Sec. II C (although he incorrectly claims that the latter is singular at the origin for the first excited harmonic oscillator eigenstate⁶¹). On the other hand, the lack of a criterion for selecting a preferred microstate implies that the quantum trajectories associated with his approach—termed “Floydian trajectories”—will not in general approach classical trajectories in the correspondence principle limits.

Actually, there is another, more fundamental reason why Floydian trajectories do not satisfy the correspondence principle. This is because the underlying dynamical law governing their evolution is radically different from that of the Bohmian quantum trajectories utilized here. The trajectories are different, *even though* the bipolar decompositions are identical—a very curious situation that bears further analysis. In particular, the period of Floydian trajectories is finite, necessitating a speed that approaches infinity as $|x| \rightarrow \infty$. In contrast, Bohmian trajectories *rapidly slow down* in the forbidden regions, never reaching the coordinate asymptotes, as discussed in Sec. IV A. Whereas Floyd finds implications for hidden variables and the Copenhagen interpretation, we adopt a decidedly less philosophically ambitious perspective.

The Floydian dynamical law is obtained from the QSHJE by applying a classical procedure,⁵² wherein trajectory evolution is related to the quantity $\partial s / \partial E$ to yield

$$\dot{x} = \left(1 - \frac{\partial u}{\partial E}\right)^{-1} \frac{1}{m} s'(x), \quad (26)$$

where $u(x) = V(x) + q(x)$ is the bipolar modified potential. Various interpretations may be provided for the $(1 - \partial u / \partial E)$ factor in Eq. (26). FM adopt a relativistic interpretation which lumps it together with m to form the “effective quantum mass,” whereas Brown (in essence) uses it to define an “effective time.” In the Bohmian approach, $u(x)$ is regarded as independent of E , thus giving rise to the usual $p = m\dot{x}$ relation. In the Floydian approach however, $u(x)$ is considered to depend on E , giving rise to the more complicated Eq. (26) expression above, for which the canonically conjugate momentum, $p(x)$, is not necessarily equal to mechanical momentum, $m\dot{x}$.

How can the same $u(x)$ be regarded as energy-independent in one theory and energy-dependent in another? It has to do with different interpretations of the energy. One can make a distinction between the quantum energy, E_Q , and the classical energy, E_C . The quantum energy is that of the eigenstate Ψ , which determines $q(x)$, $u(x)$, and $h(x, p) = p^2/2m + u(x)$. The contours of $h(x, p)$ define the classical energies E_C , with $E_Q = E_C$ corresponding to the bipolar quantum LM. The function $u(x)$ clearly depends on E_Q , but does not depend on E_C . The meaning of Eq. (26) therefore depends on whether $E = E_Q$ (Floydian approach), or $E = E_C$ (Bohmian approach).

Floydian dynamics, therefore, represents a different philosophical outlook than that of Bohmian dynamics. Both approaches are correct, and ultimately yield mathematically identical results. The main point we wish to make here is simply that this difference does not appear to have anything to do with the bipolar ansatz per se. Indeed, one could easily apply the Floydian dynamical law to the *unipolar* ansatz—although for stationary eigenstates, this would yield the same results as Bohmian propagation.

We close this section with a few final comparisons between the Floyd/FM and present approaches. First, we comment that quantum energies are discrete for bound states, and so any derivative with respect to E_Q requires careful consideration, as has been previously noted.^{51,61} This is relevant for Floydian trajectories, but is essentially a non-issue for Bohmian trajectories. Second, although Floyd has certainly considered the 1 DOF harmonic oscillator system, he appears not to have derived analytic expressions for the relevant bipolar functions, as we have done in Sec. IV. We have, moreover, verified that Floyd's closed form expression for the bipolar modified potential,⁵⁴

$$u(x) = E - 1 / \left[a\Psi_1(x)^2 + b\Psi_2(x)^2 + c\Psi_1(x)\Psi_2(x) \right]^2, \quad (27)$$

is consistent with Eq. (24), by substituting $\Psi_1 = 2r \cos(s)$, $\Psi_2 = 2r \sin(s)$, $a = b = \pi/\sqrt{2}$, $c = 0$, and $E = 1/2$ into Eq. (27). Finally, we comment that the Floyd/FM approach, being based on the QSHJE, does not appear to generalize to the non-stationary-case, whereas the present approach does—at least in certain

situations.

VI. SUMMARY AND CONCLUSIONS

The Schrödinger equation is linear, yet the equivalent Eqs. (4) and (5)—obtained via substitution of the Eq. (2) ansatz into the Schrödinger equation—are not. Quite apart from the philosophically intriguing issues which this raises, a primary conclusion of the present work is that this situation may also be exploited for practical purposes. In particular, the superposition principle allows us to divide up the initial wavepacket $\Psi(x)$ into pieces $\Psi_k(x)$, evolve each of these separately over time, and then recombine them to construct the time-evolved $\Psi(x)$ itself. So far as the Schrödinger equation is concerned, the division into $\Psi_k(x)$ pieces is arbitrary. However, if the evolution of the $\Psi_k(x)$ is performed using Eqs. (4) and (5), nonlinearity implies that the division is *not* arbitrary, but has a large impact on the time-dependent behavior of the resultant $R_k(x)$ and $S_k(x)$.

In principle, therefore, one may improve the numerical performance of quantum trajectory calculations simply by judiciously dividing up the initial wavepacket into several pieces. It remains to be seen the extent to which such a procedure will prove beneficial for actual numerical calculations of real molecular systems—to be sure, much depends on the manner in which $\Psi(x)$ is decomposed. Nevertheless, the particular bipolar scheme explored in this paper, already appears to exhibit much promise—at least with regard to ameliorating the infamous node problem, which has thus far severely limited the effectiveness of QTMs in the molecular arena.

The basic idea is to decompose a wavepacket, $\Psi(x)$, that *has* nodes, into a linear combination of two components, $\Psi_{\pm}(x)$, that do not. In practice, it is not only nodes per se that cause problems for numerical QTMs, but more generally, any large or rapid oscillations in $\Psi(x)$. Thus, $\Psi_{\pm}(x)$ should ideally be not only nodeless, but also smooth and slowly varying. For the special case where $\Psi(x)$ is a stationary Hamiltonian eigenstate, the semiclassical method is well-known to yield approximate $\Psi_{\pm}(x)$ functions with the requisite properties—even when $\Psi(x)$ itself is highly oscillatory. It is for this reason that the semiclassical solutions were used as a guide for determining the corresponding exact quantum $\Psi_{\pm}(x)$'s. Not only have we provided an explicit recipe for obtaining the latter, we have also shown that these satisfy the correspondence principle in the appropriate semiclassical limits. Thus, the bipolar quantum potential, $q(x)$, obtained here—now understood to represent the quantum correction to the semiclassical approximation—is not only well-behaved in the vicinity of nodes, but actually approaches zero, in the large action limit.

The bipolar quantum trajectories also behave very much like classical/semiclassical trajectories; indeed, the two are nearly identical in the classically allowed region, in the large action limit. This may seem paradoxical, as

both bipolar and unipolar quantum trajectories conform to the same dynamical law, and the latter are known to behave very non-classically. Unipolar trajectories do not cross in position space, for instance—which can cause kinky trajectories and other node-related difficulties, especially when the wavepacket undergoes reflection. The bipolar trajectories get around this difficulty as follows: whereas the trajectories *on a single LM sheet* never cross each other, they are all headed in the same direction anyway, and so they don't get in each other's way. On the other hand, trajectories on one LM sheet are free to cross those on the other sheet—just like the corresponding semiclassical trajectories. From a philosophical standpoint, one might thus regard the present bipolar decomposition to be more compelling than the standard unipolar approach—although curiously, this stance would require one to abandon Bohm's original pilot wave interpretation.

The above discussion anticipates future application of the present ideas to arbitrary time-evolving wavepackets; but it must be borne in mind that thus far, only stationary wavepackets have been considered. It is encouraging that the bipolar decomposition scheme outlined here was found to be preserved over time for stationary states (Sec. III C 1). On the other hand, for the more general time-evolving case, the LMs themselves will change over time, and the bipolar decomposition scheme itself need not be preserved. It is therefore possible that the initially nodeless $\Psi_{\pm}(x)$'s may develop nodes over the course of time. This need not cause difficulties in practice however, because at any desired time, one is free to redecompose $\Psi(x)$ into new $\Psi_{\pm}(x)$'s that *are* nodeless.

One of the most appealing aspects of the excited harmonic oscillator results of Sec. IV B is the fact that the bipolar functions remain smooth and slowly varying for all values of n . Indeed, the LMs for all n values resemble each other, apart from a change of scale. This is very advantageous from a numerical perspective, as it suggests that very few trajectories would be needed to accurately compute $r_n(x)$ derivatives, if a completely numerical propagation scheme were adopted. More to the point: the number of trajectories required should be essentially *independent* of the number of nodes. For sufficiently large n , it should even be possible to perform an accurate calculation with *fewer than n trajectories*—a prospect that would be virtually unheard of in a unipolar context. Note that since the bipolar trajectories themselves are also much smoother than the unipolar trajectories, far fewer time steps should be required in the bipolar case.

We conclude with a brief discussion of the prospects for multidimensional systems. At present, it is not entirely clear how best to apply bipolar decomposition to an arbitrary, real, multidimensional wavepacket, $\Psi(x_1, \dots)$. For regular systems, semiclassical theory suggests an essentially direct-product decomposition, via pairs of action-angle coordinates. Generally speaking, however, it is difficult to find these coordinates—unless the wavepacket is

initially separable, which is very often the case in molecular applications. On the other hand, the factor-of-two bifurcation applies to each degree of freedom separately, resulting in 2^D components total, where D is the number of DOFs. This is clearly undesirable for large D . A more effective strategy may be to bifurcate $\Psi(x)$ along the *reaction coordinate only*. These and other ideas will be explored in future publications.

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- * Electronic address: Bill.Poirier@ttu.edu
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